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Abstract

The study of radiation effects in complex silicate glasses has received renewed attention because of their use in special applications such as immobilization of high level nuclear wastes and fiber optics. Radiation may change the properties of these glasses by altering their electronic and atomic configurations. Electronic defects may cause absorption centers that limit their optical uses and also cause microscopic phase changes and dilatations. Atomic dislocations induced in the already disordered structure of the glasses may affect their use where heavy radiations such as alpha particles, alpha recoils, fission fragments, or accelerated ions are present. Large changes (up to 1%) in density may be caused. In some cases the radiation damage may be severe enough to affect the durability of the glass in aqueous solutions.

Recent literature concerning these and other radiation effects in silicate glasses will be reviewed.

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INTRODUCTION

In recent years, the study of the effect of radiation on silicate glasses has received new attention. This is a result of the glasses being asked to perform in environments where they will be exposed to nuclear radiation. For example, borosilicate glass is being used to immobilize nuclear waste for permanent isolation for thousands of years. Over this time period, the durability of glass should not deteriorate so that radionuclides can be released to the environment. Doses during this time period can be in excess of 10^{11} rad or 10^{19} alpha/gram from decay of alpha and beta emitters in the waste [1]. Silicate glasses are also being used as optical fibers to transmit light several meters from detectors that are in a radiation environment to analyzers that are placed a nonhazardous environment where personnel can operate safely.

The radiation damage of silicate glasses is something of a dichotomy because glasses are amorphous solids with no long range order and yet radiation effects occur as specific atomic rearrangements. The dichotomy confines our understanding primarily to the property changes resulting from radiation damage rather than the atomistic mechanisms that are responsible for them. Radiation can cause changes in many of the physical and chemical properties of glass. Examples are: the glass density can change, the reactivity of the glass to corrosion by solutions can change, stored energy can be created, the microstructure can be altered, and optical defects can form. In a paper of this size, it is impossible to review the entire field of effects of radiation of silicate glasses. Several excellent reviews have been published both on radiation effects in glasses used to immobilize nuclear waste [1-3], and in glasses used for optical fibers[4-6]. In this

review, we shall concentrate on the effects of radiation on glass density, durability, stored energy, microstructure, and optical defects.

Before discussing these subjects, it is appropriate to review the modes of interaction with silicate glasses of the various types of radiation considered in this review. Again because of space limitations, we will limit our discussion to only gamma ray and charged particle radiations such as beta and alpha particles, alpha recoils, and heavy ions. Neutrons have been used and their effects investigated. It is also instructive to give brief mention to examples of glass compositions that have been usedd to study radiation effects.

INTERACTIONS OF RADIATION WITH GLASS

Radiation can interact with both the electrons and the ttomic nuclei that make up glasses. Energy loss will be primarily by inelastic collisions, and elastic scattering. The physics of these processes has been thouroughly reviewed previoucsly[4,7]. Only a brief review will be presented here.

Gamma Rays and Beta Particles

Gamma rays and beta particles (electrons) interact primarily with the electronic system of glasses. As a result, many ionizations are produced by the secondary electrons which result from these interactions. Because of there low mass of these radiations, they induce only a few atomic displacements in the glass matrix [8,9]. Gamma irradiations are easily performed with external sources such as Co-60. Dose rates from Co-60 sources range from 10^4 to 10^7 rad/hr and doses as high as 8.5×10^{10} rads have been given the glass in laboratory tests. Electron irradiations are performed with accelerators or with electron

microscopes. The dose rate of these irradiations are on the order of 10^5 higher than the Co-60 gamma irradiations. Because of the relatively short range of the electrons, the effects are concentrated in a narrow surface layer on the glass and stresses are formed between the irradiated and nonirradiated glass.

Alpha Particles and Alpha Recoils

Alpha decay results in the formation of high energy alpha particles (~4 to 6 MeV), an energetic recoil nucleus (~0.1 MeV), and gamma radiation. Nearly all the energy of the recoil nucleus is lost through elastic collisions with the atoms of the waste glass. Each recoil can produce several thousand atomic displacements in the glass matrix. The alpha particle, because of its relatively low mass, dissipates >95% of its energy by ionization and the remainder by elastic collisions. These collisions, however, can produce several hundred atomic displacements. It has been thought that atomic displacements are the primary source for radiation effects in complex nuclear waste glasses. In these glasses, alpha decay produces several orders of magnitude more displacements than any other radiation source; thus, most studies oncerning radiation effects on nuclear waste glasses have emphasized alpha decay [2,3]. Glasses have been doped with high specific activity alpha emitting isotopes such as Pu-238 ($t_{1/2}$ = 88y) or Cm-244 ($t_{1/2}$ = 18y) to achieve the large radiation doses that nuclear wastre glasses will recevive over thousands of years of storage (Figure 1). This doping technique has been widely used to test nuclear waste glass and is the basis for the Materials Characterization Center's MCC-6P Method for Preparation and Characterization of Actinide-Doped Waste Forms [10].

Heavy Ion Radiations

Irradiations with low energy heavy ions from accelerators have been used to severely damage the surfaces of glasses. These result

in many displacements but with accompanying ionizations. Beams of many types of ions have been used. Examples are Pb ions (E = 40 - 250 keV), Xe ions (E = 160 keV) have been used. Lighter ions such as H^+, D^+, and He^{++} have also been used in studies to compare there effect with the heavier ions. These irradiations suffer from the same problem as the external electron irradiations mentioned earlier in that stresses are built up. The radiation damage is concentrated in a thin layer on the surface of the glass, and the dose rates are extremely high. Examination of the irradiated surface by such techniques as Rutherford backscattering spectrometry (RBS) and elastic recoil detection (ERD) have established that the surface of the glass is extensively damaged.

A comparison of the relative amounts of the energy of the various radiations lost in electronic and atomic processes is presented in Table I for various types of radiations. This fraction varies with the energy of the particle as it passes through the glass and its velocity decreases. The values in Table I are presented only to illustrate differences in the interactions of the radiations. Results in Table I are taken from the work of Arnold [11] and Primak [12].

EXAMPLES OF GLASS COMPOSITIONS INVESTIGATED

The elemental composition of a glass drastically affects its properties as well as its response to radiation. The more complex the composition, the more intrinstic defects in the glass structure and the more difficult it is to interpret radiation damage effects on an atomic scale. Most of the early work on radiation damage has been on fused silica. This work has formed the starting point for describing damage centers in the more complex glasses such a borosilicate and alkali borosilicate glasses. Glasses used for immobilization of nuclear waste are very complex in that they contain the nuclear waste itself which may contain high levels of

iron, aluminum, and other elements. Examples of glasses used for fundamental studies and for immobilization of nuclear waste are given in Table II.

RADIATION EFFECTS ON GLASS DENSITY

Volume Changes from Gamma and Electron Radiation

To illustrate the complexity of the effects of radiation on silicate glasses, it is useful to start with the volume changes for vitrous silica induced by gamma irradiation. For pure silica both gamma and electron irradiations cause the glass to compact and the density increases [13]. When a reactive impurity such as hydrogen is introduced into the glass matrix, the situation is reversed- the glass expands during the gamma irradiation [13]. This illustrates the complexity of radiation effects on silicate glasses. With hydrogen present, the formation of hydroxyl and hydride bonds in the glass are detected indicating that the hydrogen has reacted with free Si and SiO bond formed by the radiation. The mechanism for the expansion appears to be directly related to hydroxyl content of the silica.

Volume changes induced by gamma radiation have been examined in more complex silicate glasses [14]. These contained varying amounts alkali borates and in some cases rare earth cations. As indicated in Figure 2, these glasses compact with dose. The effect of the initial dose was greatest and was different for each of glass suggesting that the unirradiated glasses had different concentrations of intrinsic defects frozen into them. After the initial compaction, the dose dependence of further compaction was essentially the same for all the glasses. It is interesting to note that the effect of borates in the glass is opposite to the effect of the hydrogen silica. Apparently, cations cause the glass network to collapes slightly upon iradiation while hydrogen cause it to expand. In one case bonds can relax while with hydrogen, the

amount of nonbridging oxygens is being increased. It is interesting to note that there is no evidence for a saturation value for the density change even at 10^{10} rads although the curves do have a slight curvature. Glass CGW 7740 is the most complicated and shows a relative compaction of 0.5% at 10^{10}rad.

Volume changes in complex nuclear waste glasses induced by gamma rays and electrons have been measured and the change is very slight. Hall, et al.[15] simulated ionization damage from beta decay in a nuclear waste glass by iradiating with 0.5 MeV electrons at 300°C to a dose of about 2.5 x 10^{11} rad. No significant volume changes were observed. Bibler [16] reported slight compaction (<0.05%) in a simulated defense glass irradiated to 8.5 x 10^{10} rad with ^{60}Co gamma radiation. Sato et al.[17] reported swelling of about20-50% with electron irradiation in a high voltage electron microscope and only 0.2% after gamma irradiation to 10^{9} rad. Recent results of Weber [1] indicate on 0.07% swelling in MCC 76-68 after gamma irradiation to 6.6 x 10^{8} rad. It appears that volume changes from gamma irradiation of the complex nuclear waste glasses are much larger than those induced in the simpler alkali borate glasses.

Volume Changes from Alpha/Alpha Recoil Radiation

Experiments with Cm^{244} or Pu^{238} doped glasses have shown that alpha decay can cause expansion or compaction for the nuclear waste glasses. These experiments are difficult to perform due to the radioactivity of the dopants. Density changes are measured by the sink-float technique. Results taken from the reviews by Weber and Roberts[2-3] for several nuclear waste glasses developed at Pacific Northwest Laboratory (PNL) are shown in Figure 3. These results indicate that both types of volume changes saturate within the range of ± 1.2% at a dose of about 1-2 x 10^{18} alpha decays/g. Volume changes for the simulated nuclear waste glasses from the

Savannah River Plant (SRP) [16,18] are shown in Figure 4. Figure 4 also includes recent data of Weber [1] on the expansion of Defense Waste Reference Glass similar to the SRP glasses. The dose rate variation to the results in Figure 4 is 10X. The good agreement, especially for the SRP 18 and 131 glasses, indicates an absence of a dose rate effect on this property change.

The magnitude of the volume changes appears to depend on the crystallinity of the glass (Figure 3). This may be due to the fact that radiation effects on volume are much larger for crystalline solids than for glass [2-3]. Since alpha decay produces helium within the glass, the possibility of helium causing the expansion has to be addressed. Since some of the glasses compact at the same alpha decay dose where expansion is observed in other glasses, He accumulation probably does not cause the expansion. Clearly, alpha/alpha recoil radiation is more effective in producing volume changes than is gamma rays or electrons. This is probably because more displacements are caused by this alpha/alpha recoil radiation.

Volume Changes from Ion Beam Irradiations

Arnold has recently used the cantilever beam technique to measure lateral stress or volume changes induced amphorous silica and several simple borosilicate glasses [19] along with two complex nuclear waste glasses by He, Xe, or Pb ions [11]. In silica, the stress increased with fluence and then decreased with larger doses. It was found that the compaction process could best be correlated with the displacement component of the energy of the ions. For the borosilicate glasses and the nuclear waste glass there was an expansion followed by a compaction, and in both cases, the results correlated with the ionization component rather than the atomic collision component. This correlation for PNL 76-68 is shown in Figure 5. Arnold also found that, in contrast to fused silica, there is no stress relief mechanism for these glasses at higher doses although the expansion does appear to saturate.

It is clear that the mechanism for radiation induced dilitations is complicated and both negative and positive changes can occur simultaneously depending upon the glass composition. For borosilicate and nuclear waste glasses which are probably phase separated, the volume changes appear to correlate better with ionization damage rather than displacement damage. From the alpha and ion irradiation results, and from the results of Sato [17] for gamma irradiation, the volume change saturates, radiation can anneal the expansion or compaction. This phenomenon has also been observed for stored energy as will be discussed later.

RADIATION EFFECTS ON DURABILITY

Most of the experiments to determine the effect of radiation on the durability of glass have been performed on the complex nuclear waste glasses. Durability is the one of the most important properties for these types of glasses. To investigate the effect of radiation on glass durability, glasses are irradiated and then exposed to the appropriate solution for some length of time. Durability is then measured by the amount of glass dissolved. This is best determined by measuring the amounts of elements from the glass that appear in the solution or, in some cases, by measuring the weight loss of the the glass itself. The weight loss techique is complicated by the possibility of reprecipitation of species on the glass and thus it may understimate the amount of glass dissolved. It also suffers from the fact that durability is related to a small difference between two large numbers. For irradiations with ion beams, durability is measured by comparison of irradiated and unirradiated regions on the surface of the glass after the exposure have been applied.

Durability Changes from Gamma and Electron Irradiations

Both Co-60 gamma irradiations and external electron

irradiations have been used to investigate possible effects of gamma and beta radiation on the durabilities of complex nuclear waste glases. Glasses have been exposed to such irradiations and then leached in deionized water.

In one test [20], a sample of SRP 165 glass was irradiated to 1.3×10^{10} rad with Co-60 gamma radiation. Releases in deionized water at 90°C for four elements, Si, B, Li, and Na, from the glass were then measured using a standard MCC-1 leach test [21]. This leach test was developed specfically to measure the durability of the complex nuclear waste glasses. Samples of known geometric surface area are used and the ratio of the surface area to solution volume is 0.1 cm^{-1}. Results for 3 and 7 day leach tests for the irradiated and unirradiated glass are shown in Table 3. The results are excellent agreement indicating no effect of radiation on the release. Si and B are glass network-forming elements and released by dissolution of the glass. Li and Na are network modifiers and are released by both dissolution and ion exchange. Radiation has not affected either mechanism. In another Co-60 gamma irradiation test [16] a sample of SRP 131 glass was irradiated to a much larger larger dose (8.5×10^{10} rad). Samples of this glass and an unirradiated glass were then leached according to MCC-1 procedures [21] in deionized water and a brine simulating the fluid in a salt repository. Results indicated that releases based on Si, Na, B, and Li were 30% higher from the irradiated glass. The slight increase appeared real since it was consistent for all the samples.

McVay and Pederson [22] irradiated PNL glass 76-68 to a dose of 2.5×10^{8} rad and then leached it at 50°C in the MCC-1 test. Resulting solution concentrations compared to those for an unirradiated glass are in Table 4. Based on Na, Si, and B, the major elements in the glass that are completely soluble in the leachate, the irradiated glass leached 20% faster than the unirradiated glass. The concentration of the other elements were low and affected by solubility constraints; thus, they are not good

indications of the effect of radiation on the glass. It appears that either gamma radiation does not affect the durability of the glass, or if it does, the effect is slight.

Several Japanese simulated waste glasses have been irradiated with electrons to 10^{10} rad by Araki [23]. No significant increase in leach rates was observed. Bonniaud [24] observed no changes in the cesium or strontium release rates from a French waste glass after electron irradiations to 10^{11} rad. As mentioned earlier, this type of irradiation is localized in a narrow surface region of the glass and induces stresses. Even with these stresses, it appears that the glass durability is not significantly affected.

Durability Changes from Alpha Particle and Alpha Recoil Irradiation

Determination of the effect of alpha/alpha recoil radiation on the durability of glasses requires special facilities since the glasses and the resulting solutions are radioactive. Because of the presence of radioactivity, durability has mostly been determined by the weight loss technique rather than by the more sensitive solution measurements. As a result, the effect of alpha damage on glasses, although it appears to be less than 3X, has not been clearly established.

Two studies have been published where solution concentrationsa were measured. In one,the leachability of Cm-244 and K from PNL 777-260 glass has been measured [25]. In the other, PNL 76-68 glass was examined [26]. Results for the first are shown in Figure 6. Potassium is soluble in the leach solution and, thus, is a good indicator of glass dissolution. Seven Cm-244 specimens were prepared and leached in deionized water at 25 °C. The button specimens were suspended in the water, which was sampled and changed on a weekly basis for eight weeks and on a monthly basis

for three more months. The leach rate for K is also shown for an undoped glass. As can be seen, the leach rates for K and Cm-244 for the doped glasses did not change significantly with time indicating that the alpha radiation was not effecting the leachability of these two elements. The leach rates for K were significantly higher than those based on Cm-224 because of the lower solubility of Cm-244.

In the second test where solution concentrations were measured [26], samples of Cm-244 doped PNL 76-68 and 77-260 glasses were stored until they had assumulated alpha/alpha recoil doses of 3 X 10^{18} alphas/gram. A sample of each glass was annealed at 500°C for two hours to remove the damage. The leach behavior of the annealed and damaged glasses were then compared in deionized water at 90°C. No consistent differences were observed indicating no significant effect of the alpha/alpha recoil damage. As shown in Figure 2, these two glasses are not extemely sensitive to radiation induced volume changes.

The effect of internal alpha radiation on the leachability of Cm-244 and Pu-239 doped SRL 131 glass has been determined by measuring the release of Cm and Pu from the glass [16,18]. The alpha dose rate of the Cm-244 doped glass was 2.3 x 10^4 times greater than that for the Pu-239 glass because of the higher specific activity of Cm-244. In these tests the glass was placed in fresh deionized water leachant at various times. Results for both glasses in terms of the leach rate determined in each specific time increment are shown in Figure 7. Both glasses show comparable leach rates and both show a leach rate decrease as the leach time increases. The consistent decrease in the leach rate with time for both glasses indicate that they are both leaching by the same mechanism. Since the alpha dose rates in the glasses differed by a factor of 23000, there appears to be no effect of intense alpha radiation on this mechanism and no effect of dose rate on the releases. During the tests, the Cm-244 glass absorbed a dose of

1.3×10^{18} alphas per gram, and since there is no increase in Cm-244 leach rate as the radiation damage accumulates, apparently the damage is not affecting the release of Cm-244.

A number of other waste glass compositions have been doped with ^{244}Cm or ^{238}Pu for investigation of alpha decay effects. Results are summarized in Table 5. For all these tests, the effects of alpha decay on leach rates was based on weight loss in the leach tests rather than solution concentrations. The Soxhlet technique was used for the leach test. In this technique, deionized water is passed over the glass specimen in a recirculating fashion for a known time. After leaching, the change in weight of the specimen is measured. The compositions of the glasses and doping levels have been previously reported by Weber and Roberts [2,3]. The European glasses were generally doped with ^{238}Pu, as reported by Burns et al.[27] while Cm-244 was used for the PNL glasses. All these data, summarized in Table 5, have been discussed in detail by several authors [3,27,28]. The data suggest no more than a factor of 3 increase in leach rates as a result of radiation damage from alpha decay.

Recently, Weber [1] has done an interesting analysis of the data from leach studies on alpha damaged glass. He relates leach rate changes in Table 5 to the the radiation induced volume changes at the time of leaching. The analysis suggests that the largest leach rate changes are observed for those glasses which exhibit the largest volume changes and conversely, if a glass is not susceptible to alpha induced volume change, its leach rate is not affected by the radiation. Results of this analysis for several glasses are shown in Figure 8. The shape of this Figure 8 suggests that for those glasses whose volume changes are sensitive to radiation, the effect of radiation on leach rate is greatest when the effect of radiation on volume change is smallest. For example, for PNL 72-68-a glass, L/Lo varies from 1.6 to 2.6 or a factor of 1.6 while the volume decreases by less than a factor 1.1 and the

dose increases by a factor of 4. Weber [1] concludes that for some glasses, the factor of 3 may be low and leach rate changes resulting from large amounts of alpha damage may be much larger. Clearly, the values for L/Lo have to be known accurately, especially for glasses such as PNL 72-68-a to verify this relationship. It is clear that more data are needed to establish whether this correlation is correct.

Durability Changes from Ion Beam Irradiations

Irradiating the glass with heavy ions has been used to determine the effect of this type of radiation on durability. Since the resulting glass and solutions are nonradioactive, these tests are easier to perform than those the alpha doped materials. Durability measurements have employed both solution measurements and examination of the irradiated surfaces.

In those tests where solution measurements were used to measure the durability, samples of glass were exposed to beams of accelerated Pb or Xe ions. In some cases, the energy of the ions was adjusted to simulate the energy of the alpha recoils (50-250 keV). In these irradiations, very large doses can be given to the glass in a matter of seconds. The dose given to the glass is reported as ions per square centimeter of glass irradiated. The range of these ions is only 1000 A°, so the damage is concentrated on the surface of the glass. With this range, a dose of 3×10^{13} ions/cm^2 corresponds to greater than 10^{18} alpha recoils/gram glass. In this type of test, both sides of a polished sample of glass are irradiated with the ion beam and the glasses are then leached. In some cases, their surface examined with appropriate analytical techniques such as RBS and ERD. There is no question that ion beam irradiations severely damage the surface of glass. The physics of the intercction process is similar to the effect of alpha recoils in the nuclear waste glasses but the spatial distribution and dose

rates significantly different. As a result, it has been concluded the ion beam irradiations do not exactly simulate alpha recoil effects in the nuclear waste glasses [1].

In one series of tests [16] with ion beam irradiations, samples of SRP 131 glass were irradiated with 160 KeV Xe ions to two different doses, 10^{12} and 3 X 10^{13} ions/cm^2. The glasses were leached in five successive 5 hour tests in deionized water at 90°C. Very short leach times were used so that irradiated surfaces formed a large fraction of the glass leached. Leach rates based on Si, Li, and B are shown in Figure 9 as a function of cumulative leach time and dose. The rates for the unirradiated and the two irradiated glasses are in agreement indicating that the ion irradiations were not significantly affecting the durability of the glass. The irradiation is also not affecting the mechanism of the leaching process since the rates for all the glasses decrease as the leach time increases.

In another series of tests by Arnold, et al., [29] samples of SRP glass 131 were irradiated with Pb ions of varying energies (40-250 keV). This energy range was used so that a uniform damaged region could be obtained on surfaces of the glass. The glasses were irradiated on both sides to three doses. These samples were then leached in five successive tests in deionized water at 90°C. An unirradiated sample was also leached and a blank test performed. Resulting concentrations for Si, Na, B, and Li, are given in Table 6. Again, the data in Table 6 indicate no effect of radiation on the release of elements from the glass. After the fifth leach test, the surfaces of the glasses were examined by the RBS and ERD techniques to measure the relative concentrations of varous species in the surface of the glass. Results indicated that the ion irradiations caused the composition of the leached surfaces to differ. For example, components of the glass such as Li and Fe appeared more mobile in the glass surfaces that had been irradiated. After leaching, Li was more depleted and Fe more concentrated in those surfaces that had been irradiated. Even

though higher mobility was detected in the irradiated glasses, the leach rates remained unchanged. On this basis, it appears that the release of elements from the glass is controlled by the chemistry of the glass/water interface rather than mobility of species in the glass.

In a final series of tests with ion beams where solution measurements were made, samples of SRP-131 glass were irradiated with 207 KeV Pb ions to eight different doses up to a maximum dose of 10^{16} ions/cm^2 [29]. In this series, the glass was leached for very short times (1.5 hr.) for a total of 4 leach tests to ensure that only the damaged surface was being leached. Two unirradiated glasses were also leached. The concentrations of Si, B, Li, and Na in the solutions was then measured. Results for the first 1.5 hour test are given in Table 7. Within the scatter of the data, there is no effect of radiation on the durability of the glass. This was also observed for the subsequent leach tests. As with the other samples, RBS and ERD analyses of the leached samples did indicate that irradiation caused compositional differences in the leached glass.

Researchers have performed ion irradiations of other glasses and have leached these glasses in a variety of solutions ranging from 3% HF to NaCl brines. Dran, et al. [30] have given a good review of these studies. In these studies, solution concentrations were not measured. After the leach tests, the durability of the glass was estimated by measuring changes in the surface of the glass caused by leaching. Portions of the sample were masked during the irradiation; thus, the glass had irradiated and unirradiated surfaces on it when it was leached. After leaching, the durability was calculated by measuring the depth of the irradiated regions. This method is not as sensitive as measuring solution concentrations. For glasses containing only three components [31], large increases (≈50X) in leach rate were estimated in brines. Later studies on complex nuclear waste

glasses12,31,32] have shown that ion irradiation enhanced the dissolution for these waste glasses by less than a factor of 4. The reason for the difference between this result and the results based on solution analyses may be due to the different measurement techniques. Clearly, the results based on solution measurements are more applicable to release of radionuclides from nuclear waste glass.

STORED ENERGY IN IRRADIATED GLASSES

Ion irradiations and alpha decay can result in stored energy in silicate glasses. This energy is released when the glass is heated to 100-600°C. This property has been studied most for the complex nuckar waste glasses. Arnold [33] has reviwed the effects of ion irradiations and concluded that stored energy results primarily from atomic processes. For the nuclear waste glasses [2,3], the amount of energy is generally less than 150 joules per gram of glass and saturates at a dose of about 1-3 x 10^{17} alpha decays/g. Data for several nuclear waste glasses are shown in Figure 10 taken from Reference 3. The fact that a saturation value is reached indicates that radiation is able to anneal the stored energy as well as create it. Primak has suggested that the annealing process may be due to ionizations in the glass [34]. The effect of dose rate on the stored energy was investigated by comparing results with PNL 72-68 glass containing 8 wt.% Cm-244 with the same glass containing 1% Cm-244 [35]. It was found that the stored energy production and its saturation value were dependent only on the radiation dose.

Based on the results in Figure 10, the stored energy values are not capable of causing self-sustaining temperature excursions that would be of concern in nuclear waste glasses [2,3]. Since the specific heat of waste glasses is usually ≈1J/g, the maximum temperature rise that could occur from the sudden release of the stored energy is only 50 to 125°C.

RADIATION EFFECTS ON MICROSTRUCTURE

Microstructural Changes from Gamma and Electron Irradiations

Gamma and electron irradiations of complex nuclear waste glasses cause microstructural changes which have been detected by transmission electron microscopy. Recently, it has been reported [36] that gamma irradiations of simulated waste glass have resulted in significant bubble formation. The dose dependence of the bubble density for a series of SRP glasses has been determined by Howitt, et al [37] and is shown in Figure 11. The effect saturates at a dose of $\approx 10^9$ rad which is small compared to the total that the nuclear waste glasses will receive (Figure 1). The curves in Figure 11 are similar for the different glasses probably because of the closeness of their compositions. Hall et al. [15] reported microbubble formation at 200°C in a high-voltage electron microscope. Recently, electron irradiations [38-40] of simulated nuclear waste glasses have indicated significant bubble formation. Since bubble formation occurs with gamma and electron irradiation, it can probably be attributed to the ionization component of energy deposition rather than displacement. These voids are attributed to oxygen formation. A mechanism for oxygen formation has been proposed by DeNatale and Howitt [39]. The mechanism involves electron capture by the alkali ions in the glass. These cations then diffuse far enough from the liberated oxygen to prent geminate recombination. The liberated oxygen atoms eventually form oxygen molecules in the microstructure of the glass.

Microstructural Changes from Alpha/Alpha Recoil Irradiations

Weber, et al. investigated the microstructural changes in Cm-244 glass using x-ray diffractilon[3]. Radiation induced devitrification was not observed. Bubble formation in an actinide-doped glass was observed by Bibler and Kelley [18] after exposure to the electron beam in an SEM and was attributed to the beam-induced agglomeration of helium generated from alpha decay, since similar behavior could not be induced in an undoped glass. De, et al.[40] implanted 50 keV helium into a simulated borosilicate glass and observed bubble formation and surface blistering after heating to 600°C. This was also probably due to He agglomeration.

Microstructural Changes from Ion Irradiations

Microstructural changes caused by irradiating PNL-76-68 glass with He, Ar, or Pb ions has also been investigated by DeNatale, et al. [41]. After the irradiations, the samples were thinned for transmission electron microscopy from the unirradiated side to ensure that the irradiated surface was not damaged by the thinning process. Porous microstructures similar to those formed by gamma and electron irradations were observed. These structures were attributed to bubble formation. Based on the size and distribution of the bubbles, it was concluded that the formation was primarily due to the electronic component of the radiation and not the displacement component. An assessment of the importance of ionization damage to the immobilization of nuclear waste in glass has been given by DeNatale and Howitt [42].

RADIATION EFFECTS ON OPTICAL PROPERTIES

The optical clarity of silicate glasses is extremely sensitive to the extent of radiation damage that has occurred. Defect centers are formed that are strongly absorbing in the visible and

infra red. As a result, certain glasses have been proposed as solid state radiation dosimeters[43]. Several types of defects in amphorous silica and in alkali silicates have been identified by electron spin resonance techniques. Because excellent reviews on this subject already exist[4-6], only a brief discussion of the effect will be included here.

Point Defect Centers

The most common defect center caused by radiation in silicate glasses is designated by the E' center or oxygen vacancy. Several E' variants have been identifed and their structures proposed [44]. The E' center is is usually described as an electron trapped at an oxygen vacancy on one of the silicon atoms. This atom relaxes to the planar sp^2 configuration, which is a lower energy state, while the other remains as an sp^3. The E' center can act as an electron trap to form a distorted Si-Si bond. Three other varient E' centers have recently been identified in pure silica based on their production and thermal bleaching behavior [44]. The formation of peroxide radicals at an E' is possible by the reaction of a difusing O_2 molecule with center. Two other types of defects that can be formed in amorphous silicates is the oxygen related hole center (ORHC) and the non-bridging oxygen hole center (NBOHC). The NBOHC is, for example, a hole trapped by a broken Si-O-Si bridge.

A multitude of different defects are introduced in silicate glasses by the addition of alkali metals and network modifiers. In alkali silicate glasses, centers designated a HC1 and HC2 are formed and described as trapped holes[45]. HC1 and HC2 centers, for example, represent the non-bridging -O-Na linkage [46]. The HC1 center involves one sodium atom and the HC2 is associated with two. The HC1 is favored over the HC2 at low alkali concentrations and the situation is reversed as the alkali concentration increases. Three other types of oxygen-related defects in alkali silicate glasses have been identified[45]. These are interstitial O_2^- ions,

O_2^- ions bonded to the glass network and interstitial ozonide(O_3^-) ions. Information concerning defect centers and their anihilation in alkali silicate glasses has also been obtained from pulse radiolysis studies which detect short lived defect centers in the glass. Maekawa, et al [47] have detected defect centers which were electrons trapped on the alkali ions. The formation of these increases as the alkali content of the glass increases. The absorption spectra from these centers resembles those observed by Mackey [48] from low temperature X-ray irradiation similar glasses. On a microsecond time scale, these centers decay decay and give an absorption spectra which resembles the permanent one obtained by X-ray irradiation. Recombination mechanisms based on migration of trapped electrons in the glass has been invesitgated by Barkatt and coworkers [49]. When electron scavenaging dopants were added to the glass, the kinetics of anihilation decreased due to the slower diffusion of the electron traps.

Different types of E' centers can be annealed out of amorphous Si and alkali silicates at different tempratures. Griscom [4,44] has recently related these annealing processes with the effective temperature ranges for diffision of atomic or molecular species in the fused silica. As a result, he proposes that some of the annealing processes may be due to diffusion limited reactions with radiolytically formed H_2 and O_2.

Microscopic and Macroscopic Rearrangements

The question now arises whether the microscopic defects can aggregate to form observable phase changes. Defects could possibly aggregrate quite readily in silicate glasses at room temperature because of the high diffusivities associated with these open structures. The application of transmission microscopy to these materials has been applied to identify these phase changes. This application is hindered because the electron beam of the instrument

itself can induce the radiation effects. For example, the defect aggregation in quartz as it becomes amorphous from the electron bombardment of the imaging beam has been observed by [50]). The topographic freedom provided by the oxygen vacancies appears to be the key to the metamict transformation in quartz and the overall loss of crystallinity occurs as a parallel process to the aggregation of the defect centers. The situation is further complicated by the fact that quartz can be driven to an amorphous state by both direct displacement and radiolytic processes, that is to say from electronic and nuclear interactions. In the electron irradiations the radiolytic process dominates and experiments which combine electron and ion irradiations have shown that both types of damage give rise to the same structural decomposition [36,39].

The conclusions from the studies in silica are appropriate to the understanding of the behavior of the silica network but as the ionicity of the glasses increase, from the addition of alkali and alkaline earth cations a second, more efficient, radiolytic process apparently takes over [39]. In this case it is the activity on the cation sublattice that controls the radiolytic decomposition, even though it is again the oxygen defect that is most straightforward to detect. The mechanism to describe the behavior of alkali borosilicate glasses was alluded to earlier in this review. It was first proposed by DeNatale et al, [ ] and involves the creation of free oxygen, the migration of the associated cations and the subsequent formation of oxygen bubbles. The kinetics of the aggregation processes which ensue and subsequently stabilize the radiation damage, are controlled by the diffusion rate of the cations and that of the molecular oxygen. The oxygen vacancies may be stabilized into oxygen bubbles if the associated cation can move away and if the molecular oxygen remains trapped as the pressure within the bubble continues to increase. In the binary silicate glasses it has been shown that the precipitation of oxygen bubbles increases proportionately to the magnitude of the cation diffusion coefficient[39]. Also, bubbles will not form in thin films where the molecular oxygen can escape. In this case the outgassing of

oxygen is observed from the surfaces [51].

The response of the silicate glasses to irradiation at low dose, such as the behavior observed in optical fibers [52] depends critically upon the alkali content. The ability to remove radiation induced defects by annealing may depend very much on the extent of molecular oxygen precipitation that has occurred. The annealing of the glasses at very high temperatures has been shown to remove the oxygen defects in thin foils [51], but not in bulk specimens. It is suggested that the formation of molecular oxygen bubbles that may be responsible for the difficulty in annealing out some of the absorption bands in optical fibers. Clearly more work has to be performed to test this hypothesis.

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Table I

Relative Energy Loss Properties of Various Radiations

Radiation	Initial Energy (KeV)	Fractional Energy Loss	
		Electronic Processes	Atomic Processes
Co-60 Gamma	1200	0.99	0.01
Electrons	1000	0.98	0.02
Alpha Particles	5000	0.95	0.05
D-Ions	20	0.93	0.07
He-Ions	20	0.79	0.21
Ne-Ions	100	0.47	0.53
Xe-Ions	320	0.11	0.89
Pb-Ions	250	0.28	0.72

Taken from References 11 and 12.

Table II

Major Oxide Components of Typical Glass
Used to Study Radiation Effects

Oxide Wt %	Simple Glasses		Nuclear Waste Glasses				
	CGW 7740	Boro-Silicate	SRP 165	SRP 7131	PNL 76-68	PNL 72-68	DWRG
SiO_2	80.8	75	54.5	42.8	40.8	27.3	50.6
B_2O_3	12.0	20	6.2	10.6	8.8	11.2	7.0
Al_2O_3	2		4.4	2.7	0.6	0	6.0
CaO	0.2		1.4	1.0	2.4	1.5	2.0
Fe_2O_3			12.3	13.2	9.4	0.95	10.0
K_2O	0.6		-	0.13	--	4.1	--
Li_2O			4.8	4.1	4.1	0	4.9
MgO	0.2		0.7	1.4	0.2	1.5	0.7
MnO_2			2.6	3.8	0.06	0	3.0
Na_2O	4.2	5	10.9	13.8	11.5	4.1	9.1
NiO			0.9	1.6	0.3	0.7	2.0
ZnO			--		4.7	21.3	

Table III

Effect of Ionizing Radiation from Co-60 on Releases from SRP 165 Waste Glass[a]

	Normalized Mass Losses (g/m^2)			
3 Day Leach Test[b]	Si	Na	Li	B
Unirradiated Glass	4.5	3.8	4.7	5.0
Irradiated Glass	4.4	3.7	4.6	4.8
7 Day Leach Tests[c]	Si	Na	Li	B
Unirradiated Glass	7.2±0.3	6.6±0.3	8.3±0.4	8.4±0.4
Irradiated Glass	7.3±0.2	6.7±0.3	8.5±0.3	8.5±0.3

Taken from Reference 20.

Table IV

Effect of Gamma Irradiation on Leaching of 76-68 Glass in Deionized Water at 50°C

Element	Concentrations after Leaching (mg/L) Unirradiated	Irradiated[a]
Na	0.82	0.96
Cs	0.10	0.10
Ca	0.11	0.13
Ba	0.02	0.03
Sr	0.02	0.03
Si	1.6	1.9
B	0.26	0.29
Mo	0.10	0.12
Zn	0.06	0.07
Nd	<0.02	<0.02
Ce	<0.04	<0.04
Fe	<0.005	<0.005
solution pH before/after	5.7/7.2	5.7/7.2

Taken from Reference 22

Table V

Summary of Leach Rate Changes in Borosilicate Waste Glasses Based on Weight Loss in Soxhlet Tests

Glass	Storage Temp. (°C)	Maximum Dose (alpha decays/g)	Factorial Leach Rate Change
PNL 77-260 (Vitreous)	23	1.4×10^{18}	-1.1
PNL 77-260 (Small Crystals)	23	1.4×10^{18}	-1.3
PNL 77-260 (Large Crystals)	23	1.4×10^{18}	-5.0
PNL 72-68-a	23	3.1×10^{18}	2.0
UK 189	20[a]	5.6×10^{18}	2.5
UK 189	20[b]	5.6×10^{18}	2.1
UK 189	23	1.1×10^{18}	1.4
UK 189	170	1.1×10^{18}	1.6
UK 209	23	1.1×10^{18}	1.1
UK 209	170	1.1×10^{18}	1.2
SDN 58.30.20.U2	23	1.1×10^{18}	3.1
SDN 58.30.20.U2	170	1.1×10^{18}	1.6
VG 98/3	23	1.1×10^{18}	1.3
VG 98/3	170	1.1×10^{18}	1.1

Taken from References 2 and 3. See References therein for original data.

Table VI

Leaching Si, Na, B, and Li from SRL-131 Glass Irradiated with 250 keV Pb Ions.

Element Leached	Radiation Dose ions/cm^2,[c]	Concentration (PPM) in Leach Test[b] Test 1	Test 2	Test 3	Test 4	Test 5
Si	Blank	0.03	0.05	0.10	0.11	0.11
	0	0.16	0.28	0.20	0.26	0.35
	10^{12}	0.30	0.38	0.31	0.48	0.50
	10^{13}	0.23	0.23	0.20	0.27	0.30
	3×10^{13}	0.24	0.36	0.26	0.30	0.42
Na	Blank	0.12	0.03	0.08	0.04	0.04
	0	0.42	0.19	0.16	0.15	0.13
	10^{12}	0.41	0.15	0.25	0.16	0.23
	10^{13}	0.32	0.34	0.21	0.11	0.29
	3×10^{13}	0.39	0.22	0.24	0.14	0.16
B	Blank	0.003	<0.002	<0.002	<0.002	<0.002
	0	0.080	0.015	0.021	0.021	0.019
	10^{12}	0.083	0.020	0.031	0.047	0.044
	10^{13}	0.076	0.027	0.021	0.014	0.011
	3×10^{13}	0.085	0.016	0.021	0.016	0.019
Li	Blank	<0.002	<0.002	<0.002	<0.002	<0.002
	0	0.036	0.010	0.010	0.003	0.016
	10^{12}	0.038	0.013	0.014	0.021	0.028
	10^{13}	0.036	0.008	0.010	0.009	0.023
	3×10^{13}	0.039	0.011	0.011	0.007	0.015

Taken from Reference 29

Table VII

Leaching Si, B, Na, and Li from SRL-131 Glass
Irradiated with 207 keV Pb Ions

Radiation Dose ions/cm^2	Concentration (PPM) in Leach Test Si	B	Na	Li
0	0.126	0.030	0.183	0.028
0	0.110	0.031	0.109	0.025
5×10^{11}	0.087	0.029	0.204	0.032
10^{12}	0.032	0.023	0.124	0.026
5×10^{12}	0.079	0.026	0.158	0.028
10^{13}	0.127	0.033	0.167	0.032
5×10^{13}	0.109	0.029	0.210	0.029
10^{14}	0.046	0.030	0.171	0.026
10^{15}	0.162	0.042	0.187	0.037
10^{16}	0.187	0.039	0.221	0.041

Taken from Reference 29.

Table 8

Leaching Si, B, Na, and Li from SRL-131 Glass
Irradiated with 207 keV Pb Ions

Radiation Dose ions/cm^2	Concentration (PPM) in Leach Test			
	Si	B	Na	Li
0	0.126	0.030	0.183	0.028
0	0.110	0.031	0.109	0.025
5×10^{11}	0.087	0.029	0.204	0.032
10^{12}	0.032	0.023	0.124	0.026
5×10^{12}	0.079	0.026	0.158	0.028
10^{13}	0.127	0.033	0.167	0.032
5×10^{13}	0.109	0.029	0.210	0.029
10^{14}	0.046	0.030	0.171	0.026
10^{15}	0.162	0.042	0.187	0.037
10^{16}	0.187	0.039	0.221	0.041

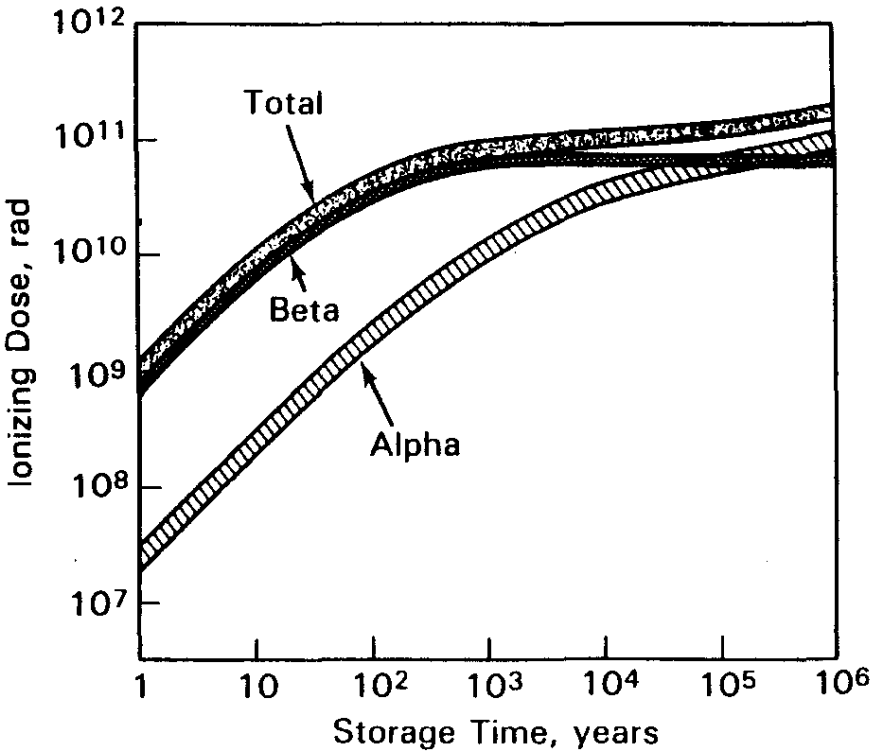

Figure 1. Calculated Radiation Doses to Defense Nuclear Waste Glass (Reference 1)

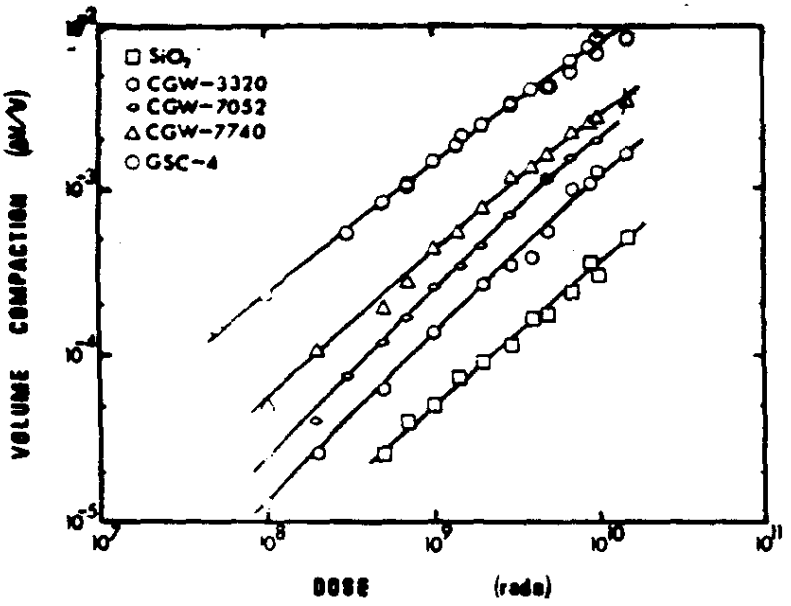

Figure 2. Gamma Ray Induced Compaction in Fused Silica and Borosilicate Glasses (Reference 14)

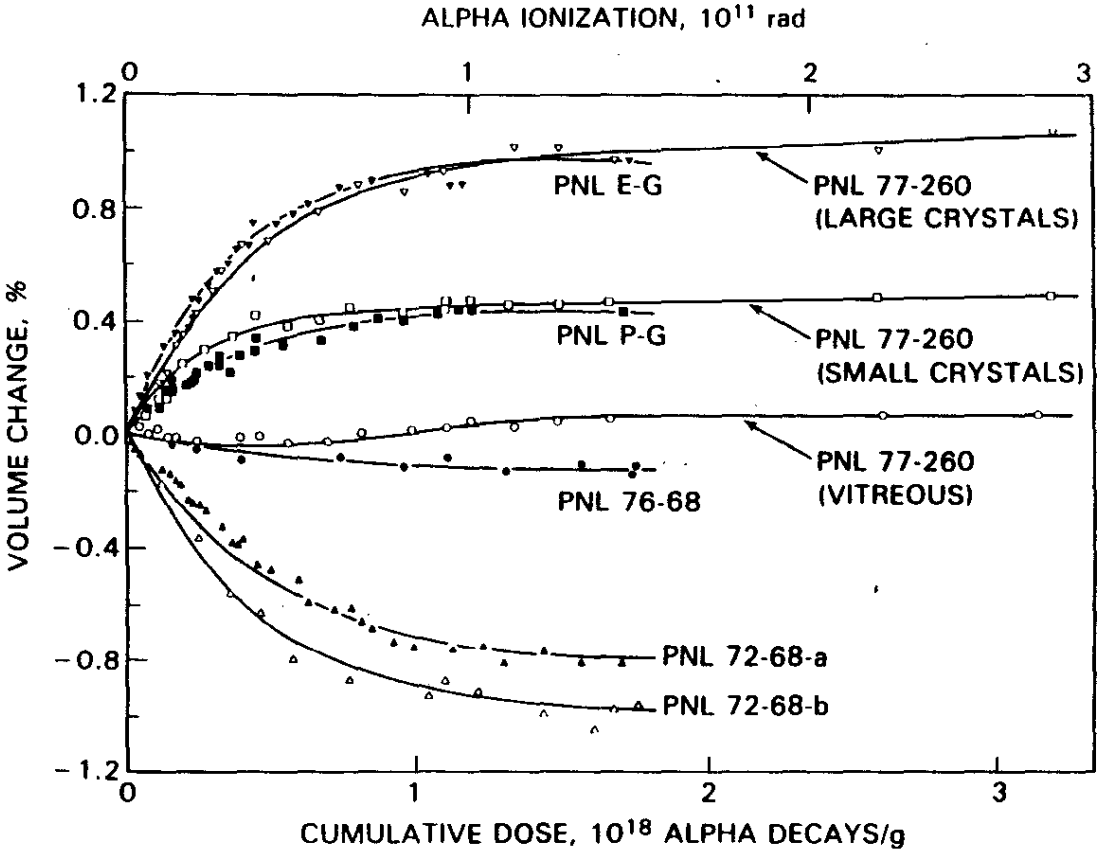

Figure 3. Alpha Particle and Alpha Recoil Induced Volume Changes in Pacific Northwest Laboratory Nuclear Waste Glasses (Reference 3)

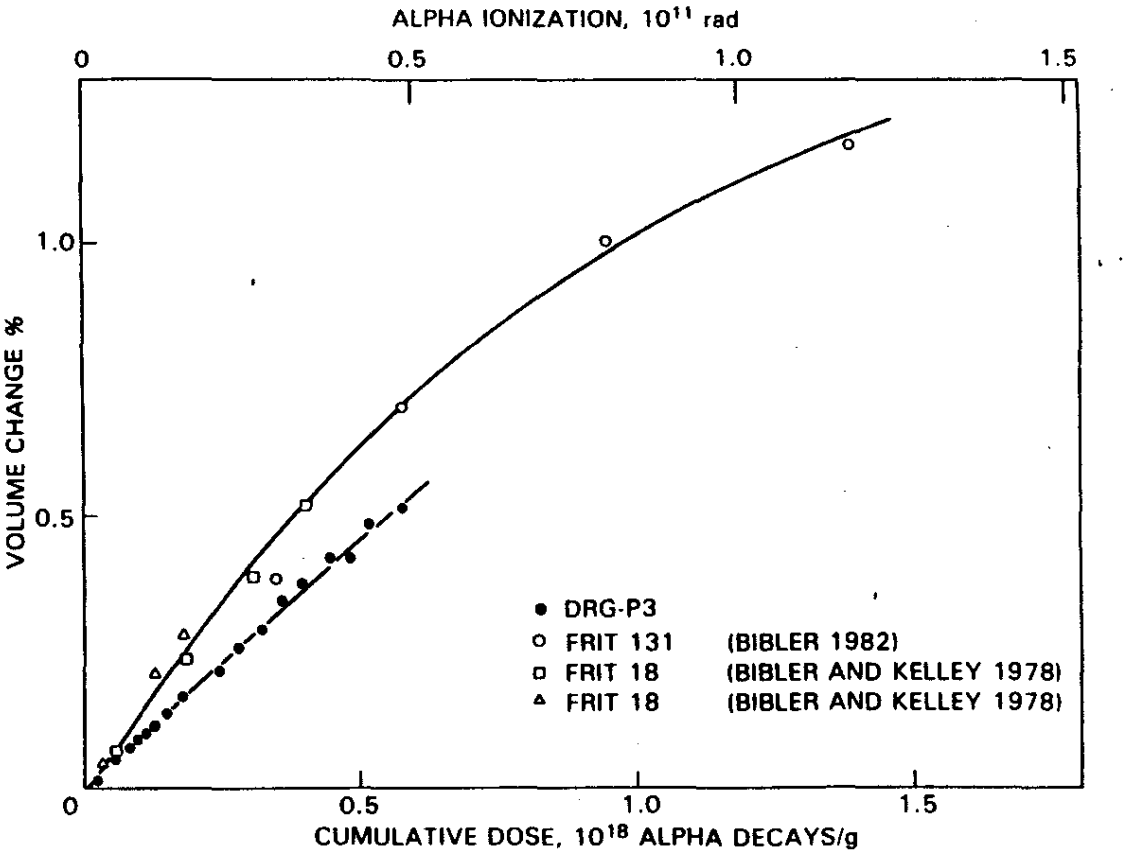

Figure 4. Alpha Particle and Alpha Recoil Induced Volume Changes in Savannah River Plant Nuclear Waste Glasses (Reference 16 and 1)

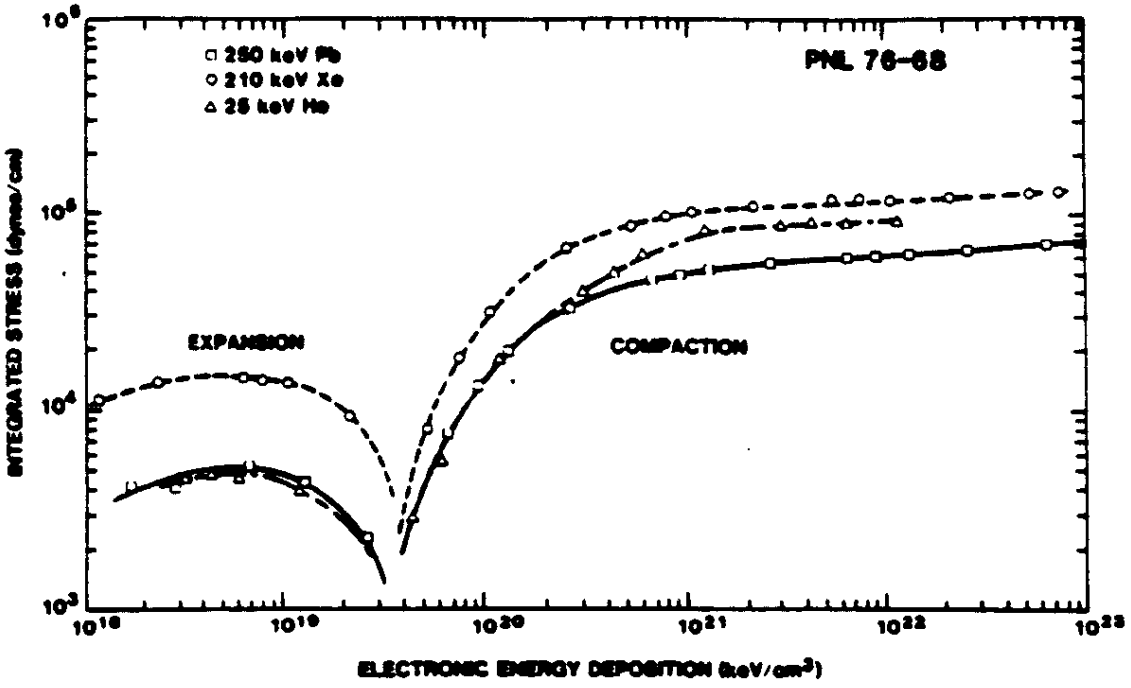

Figure 5. Volume Changes Induced in PNL 76-68 Glass by Iradiation with Pb, Xe, or He Ions. Integrated stress caused by the volume change is plotted as a function of the electronic component of the energy deposition. (Reference 11)

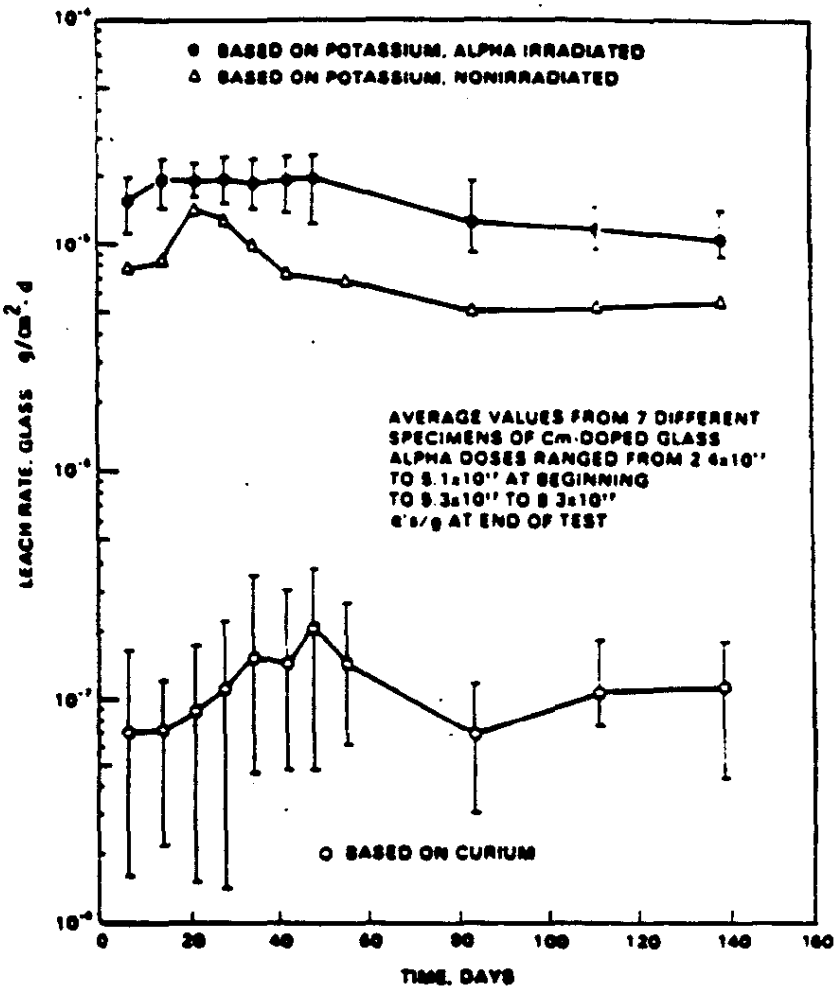

Figure 6. Effect of Internal Alpha Radiation on Leaching K and Cm-244 from PNL-72-68 Glass (Reference 25)

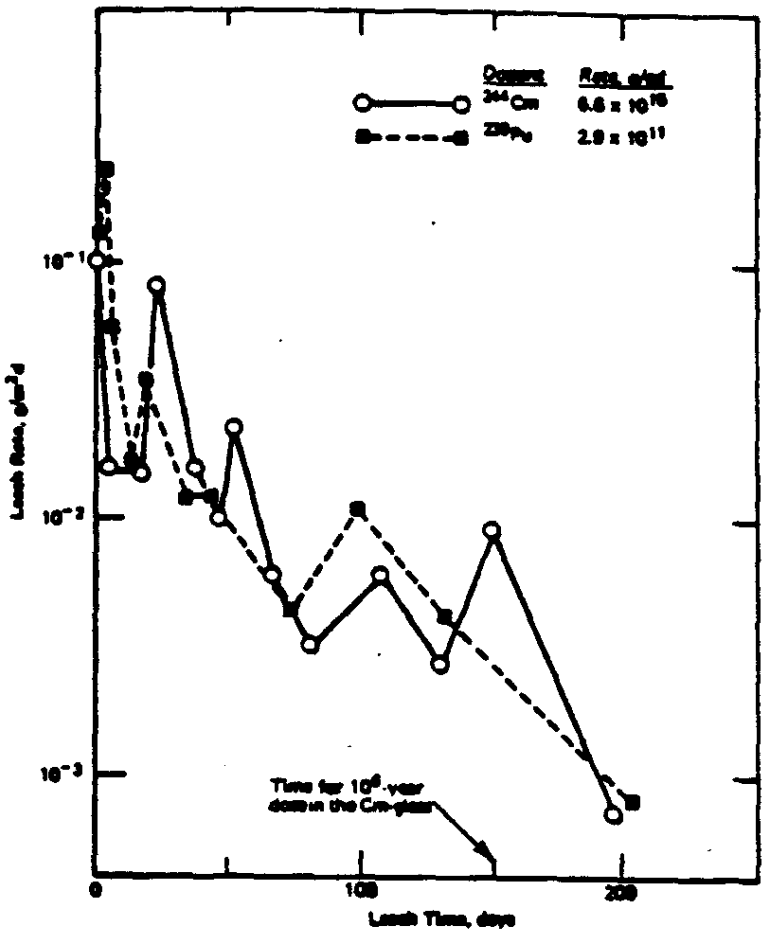

Figure 7. Comparison of Leach Rates for Cm-244 and Pu-239 Doped SRP 131 Glass. Alpha dose rate to the Cm-244 glass is 23,000 times larger than that to the Pu-239 glass. (Reference 16)

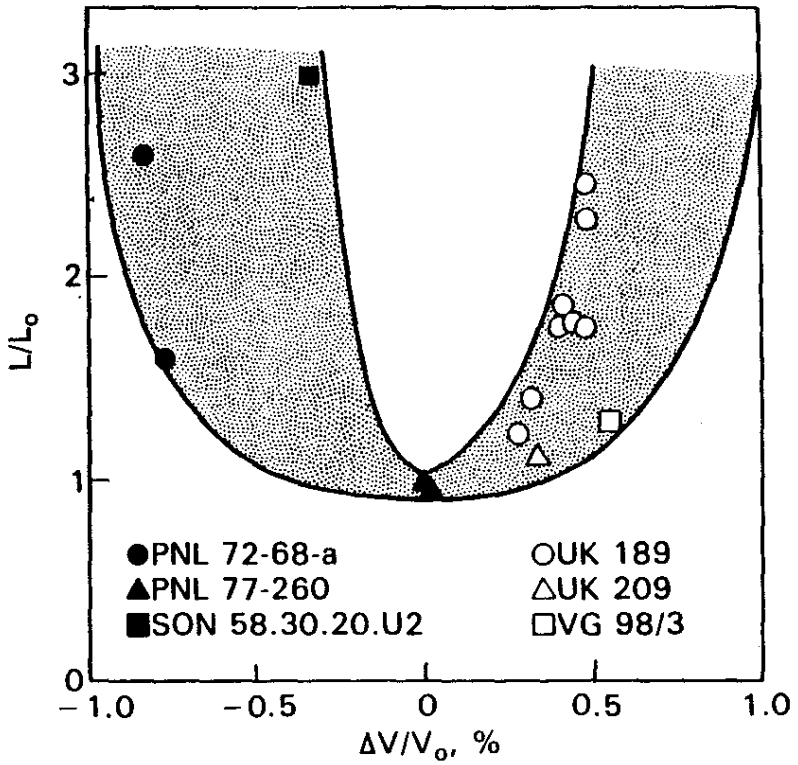

Figure 8. Effect of Alpha Induced Volume Change on Leach Rate Change for Several Nuclear Waste Glasses (Reference 1)

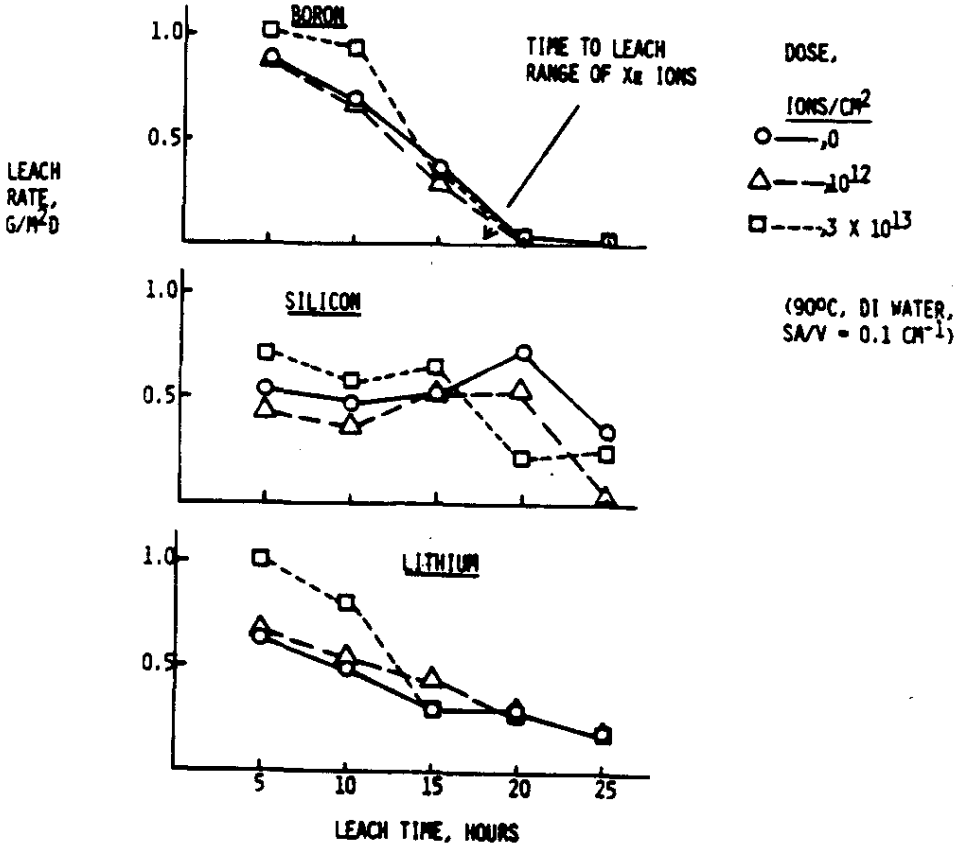

Figure 9. Effect of 160 XE KeV Ion Irradiation on Leaching SRP 131 Glass in Five Successive Leach Tests in Deionized Water at 90°C. (Reference 16)

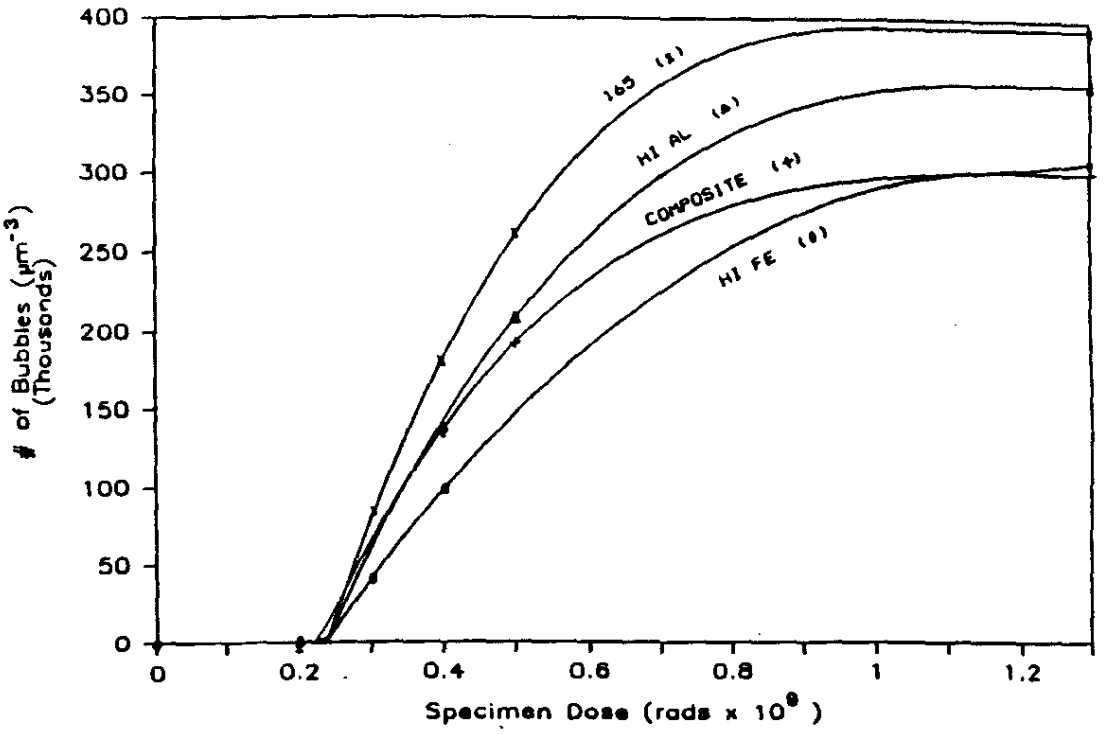

Figure 10. Density of Bubble Formation from Gamma Ray Irradiation of Four SRP Glasses Determined by Transmission Electron Microscopy (Reference 37)

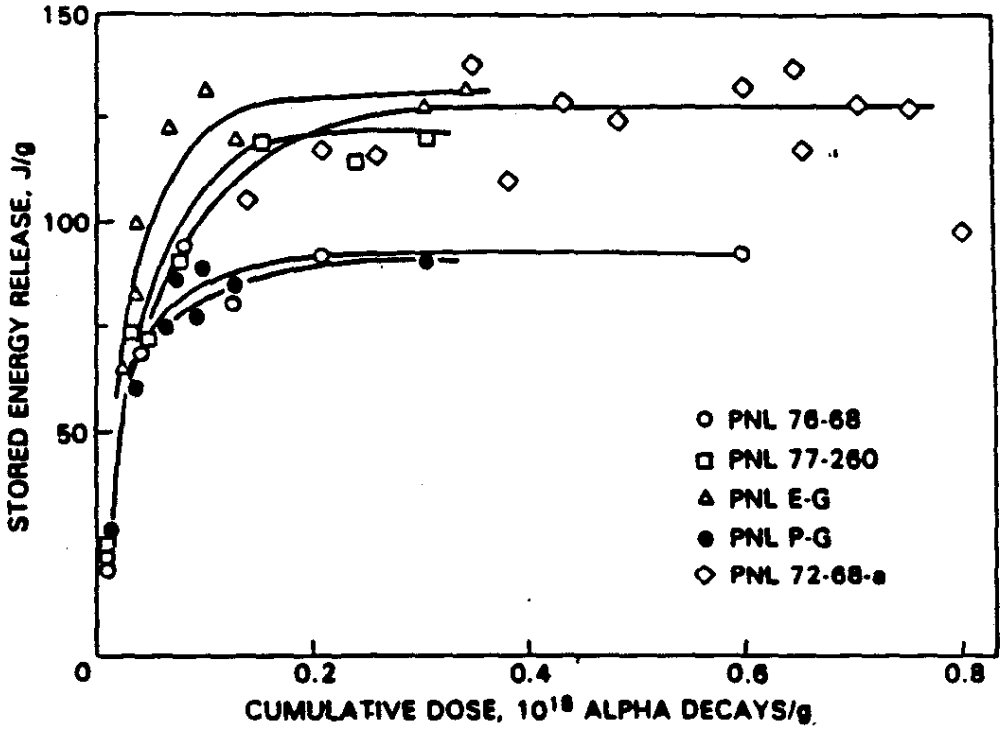

Figure 11. Stored Energy as a Results of Alpha Decay in Several PNL Simulated Nuclear Waste Glasses (Reference 4)